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## Fiber charge characteristics of pulp suspension containing aluminum sulfate

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**Abstract** Fiber charge characteristics of pulp suspensions containing aluminum sulfate were investigated with relation to adsorption behavior of aluminum components on the pulp fibers by streaming potential measurement using a particle charge detector, X-ray photoelectron spectroscopy, and X-ray fluorescence analysis. When aluminum sulfate was added to a pulp suspension prepared using deionized water, a streaming potential of the suspension went from negative to slightly negative according to the adsorption of aluminum components on the pulp fibers. Subsequent addition of a dilute NaOH solution to the suspension drastically cationized the fibers in the pH range of around 5 by predominant and homogeneous adsorption of cationic aluminum complexes on the fiber surfaces. However, the aluminum flocs that formed heterogeneously on the fiber surfaces at higher pH by further alkali addition made nearly no contribution to cationization of the fibers, although the abundant aluminum components were retained in the pulp sheets. Therefore, only aluminum cations adsorbed uniformly on the fiber surfaces perform well to control the charge properties of the pulp fibers at the wet end; and the preferential aluminum adsorption behavior on the fiber surfaces, by utilizing the required amounts of hydroxyl ions, probably accounts for the effective cationization of the fibers under acidic to neutral papermaking conditions.

**Key words** Fiber charge · Aluminum sulfate · Streaming potential · X-ray photoelectron spectroscopy · X-ray fluorescence analysis

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### Introduction

Electrostatic charges of dissolved and suspended materials in paper furnish directly influence the retention behavior of fines, fillers, and various wet-end additives that improve paper quality and the process runnability. Aluminum compounds such as aluminum sulfate, commonly called alum, have been widely applied to practical papermaking processes to control the charge properties of the anionic substances at the wet end, and they have thus performed as drainage and retention aids. Structural and electrostatic properties of Al compounds in aqueous media, however, are susceptible to the pH of the system, its temperature, the shearing forces in the liquid flow, and other conditions.<sup>1-5</sup> Therefore, effective application in the practical processes has long been studied with regard to the adsorption behavior of a variety of Al compounds on the pulp fibers at the wet end<sup>3,6-11</sup> and the electrostatic variation with partial polymerization of Al species by olation and oxolation in pure aqueous systems.<sup>2,4-7,12</sup> It has been reported that the cationic Al oligomer is stoichiometrically produced by moderate hydrolysis of trivalent Al ions,<sup>4</sup> and Al cationicity is governed by the alkalinity of the Al solutions.<sup>2,5,12</sup> Moreover, Al retention increases with increasing pH of the pulp suspension by adding alkali solution at around pH 5.<sup>6</sup> However, almost no close relations among the formation of Al-polycation complexes, Al cationicity in aqueous media, and Al content of the paper sheets were confirmed at pH 4–6 of each system, as roughly summarized in these reports. One of the reasons is that the retention behavior of Al components on the pulp fibers at the wet end must depend on several mechanisms (i.e., electrostatic interactions and simple entrapment by filtration effects).<sup>9-11</sup> Moreover, charge properties of Al-adsorbed insoluble substances such as fibers and fines have not been studied in detail. Meaningfully, the accessibility of Al ions to the cellulosic pulp fibers must be different from that of other wet-end constituents such as fines, fillers, and colloidal agents because the Al ions are smaller than these substances. Thus, the superficial regions of the pulp fibers accessible for the

larger substances in the paper stock were investigated to clarify the complicated drainage and retention mechanisms and to improve the efficiency of Al compound usage. However, many difficulties have arisen in the determinations with regard to the adsorption behavior of the Al components and their concrete performance at the wet end.

The ionic interaction between various constituents at the wet end is of significance for effective flocculation and dispersion. Their charge properties in colloidal systems have long been determined accurately by polyelectrolyte titration or  $\zeta$ -potential measurement.<sup>13–18</sup> Recently, streaming potential measuring devices, instead of metachromophoric indicators such as *o*-toluidine blue, have been reported as on-line detectors of accurate endpoints during conventional colloid titration using polyelectrolyte reagents.<sup>19</sup> It was also reported that the streaming potential values of the suspensions containing polyelectrolytes or some inorganic particles corresponded quite well with their  $\zeta$  potentials.<sup>20,21</sup> Thus, the use of representative streaming potential measurements using a particle charge detector (PCD), which mainly detects the surface charges of suspended particles and colloids in the systems, has been attempted for practical application as an easy-handling, accurate analytical technique to detect charge properties in various colloidal systems.

Quantitative analyses of Al components present in paper sheets are indispensable for investigating Al adsorption behavior on pulp fibers at the wet end. A variety of analytical techniques have been adopted so far [e.g., atomic absorption spectroscopy, energy dispersive X-ray analysis (EDXA),<sup>9,22</sup> and X-ray fluorescence analysis (XFA)].<sup>9–11,23</sup> These methods can determine almost all of the detectable elements in the specimens to some extent in each principle. In comparison, X-ray photoelectron spectroscopy (XPS) has been available for analysis of the outermost surface layers of various metallic and inorganic materials and has recently been applied to organic substances.<sup>24–26</sup> It is noteworthy that the detectable regions measured by XPS are quite shallow (< about 10 nm in depth), and it is possible to obtain the particular information restricted to the surface characteristics. Furthermore, the mapping images of plural elements on the sample surfaces can be observed simultaneously by a scanning analytical mode using a split field device for photoelectron detection.

In this study, streaming potentials of the pulp suspensions containing aluminum sulfate with various degrees of alkalinity, the relative amounts of an Al element present only on the fiber surfaces, and the Al content of the pulp sheets were determined by the PCD system, XPS, and XFA, respectively. Two-dimensional XPS analysis was applied to evaluate the distribution characteristics of Al components on the sheet surfaces. Then the surface charge properties of the pulp fibers at the various pHs of the suspensions were investigated with regard to adsorption behavior of Al components on the pulp fibers estimated by the quantitative and dispersive differences of the Al components on the fiber surfaces.

## Materials and methods

### Materials

A commercial bleached hardwood kraft pulp was sufficiently disintegrated in tap water and beaten to 450 ml of Canadian Standard Freeness<sup>27</sup> using a laboratory-scale beater with a controlled bedplate.<sup>28</sup> The pulp obtained was soaked in 0.1 M NaOH solution at 20°C for 2 h and then washed thoroughly with deionized water. Subsequently, the pulp was soaked in 0.1 M HCl solution at 20°C for 2 h followed by similar washing to remove anionic contaminants and ionic materials preadsorbed on the pulp.

The carboxyl content of the pulp was 0.06 mmol/g of the dry pulp determined by acid-base back-titration according to a conventional standard method.<sup>29</sup> An anionic rosin emulsion size (AL-120; Japan PMC, Japan) was a commercial product. Aluminum sulfate, potassium polyvinyl sulfate (KPVS), polydiallyldimethylammonium chloride (PDADMAC), and other chemicals were reagent grade (Wako, Japan) and were used without further purification.

### Preparation of sheet samples

Pulp suspensions with 0.15% consistency by weight per volume were prepared with the beaten pulp using deionized water; a constant amount of aluminum sulfate (2% on dry pulp) was added to the suspensions, whose pH values and alkalinity were controlled by the following addition of a dilute NaOH solution during the streaming potential and pH measurements of the suspensions. Anionic rosin emulsions (1% on dry pulp) as typical ionic colloidal particles were finally added to part of the pulp mixtures before filtration for sheet formation. These pulp mixtures with or without rosin sizes were then filtered using 200 mesh wire, and the wet-pressed web pads were dried without restraint at 20°C and 65% relative humidity for more than 24 h. These procedures were carried out according to a standard method for handsheet manufacture.<sup>30</sup> The pulp sheets with rosin sizes were applied to pyrolysis-gas chromatography (Py-GC) for determining the rosin content of the sheets. The pulp sheets without rosin sizes were subjected to both XPS and XFA to quantify the Al components in the sheets.

### Streaming potential and pH measurements

Streaming potentials of the pulp suspensions containing aluminum sulfate were consecutively measured using a PCD equipped with a potentiometric titrator having auto bullets (PCD-100; Kyoto Electronics/Mütek, Japan).<sup>20</sup> The pulp suspension prepared in the previous section was poured into the nonconducting cylindrical vessel of the PCD; and a piston coated with polytetrafluoroethylene moved vertically in the vessel. The reciprocal motion (4 Hz) of the piston with 5 mm amplitude caused rapid flow of the pulp suspension in the opposite direction within the annular

space. Some of the pulp fibers had interactions with the vessel walls; and the diffused electrical double layers of the fibers were distorted by the liquid flows. The electrical difference was measured with electrodes of the PCD as the AC signal, and the continuous changes in the signals were monitored on-line while the aluminum sulfate and alkali solutions were added to the medium. The pH values of the pulp mixtures were measured simultaneously using a standard pH meter.

### XPS analyses

Aluminum components present on the fiber surfaces were determined quantitatively using an XPS (AXIS-HSi; Shimadzu/Kratos, Japan) equipped with a low-energy electron flood gun as a neutralizer to correct the positive charges due to photoelectrons escaping from the surfaces of the insulating specimen during XPS measurement.  $MgK\alpha$  and monochromatized  $AlK\alpha$  radiations were used to excite the photoelectrons from the sheet samples for two-dimensional and spectroscopic XPS analyses, respectively. X-ray generation conditions were 15 kV and 10 mA. These analyses were carried out in the low pressure ( $0.2\mu Pa$ ) range at least three times on different locations of the specimen, with each area being about  $0.5\text{ mm}^2$ , for quantification. The mapping area was divided into 1024 regions of  $0.2 \times 0.2\text{ mm}$  and was scanned horizontally using a split-field apparatus of the XPS system. Measuring conditions in narrow scans for high-resolution determination were 10 eV of pass energy and 0.05 eV of step width; 80 eV of pass energy in carbon and Al elements was available for the mapping scan. Binding energies (BEs) of all spectra obtained were related to a conspicuous carbon 1s signal (unoxidized C—C band) at 285 eV.<sup>24-26</sup>

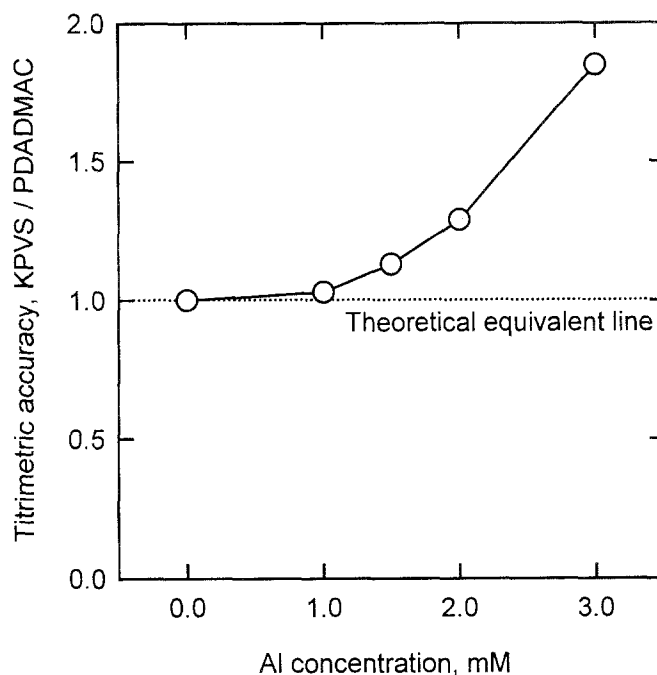
### Other analyses

Aluminum content of the pulp sheets was determined using an XFA (MESA-500; Horiba, Japan) with a quantitative analysis program.<sup>9,23</sup> X-ray generation conditions were 15 kV and 0.5 mA, and the irradiation time was set to 100 s. The analysis was performed at least three times at different locations on the specimen. Rosin size content of the sheets was determined by Py-GC combined with the on-line methylation method using a 25% tetramethylammonium hydroxide-methanol solution.<sup>23</sup>

## Results and discussion

### Titrimetric accuracy of PCD system for colloid titration of Al-containing pulp suspension

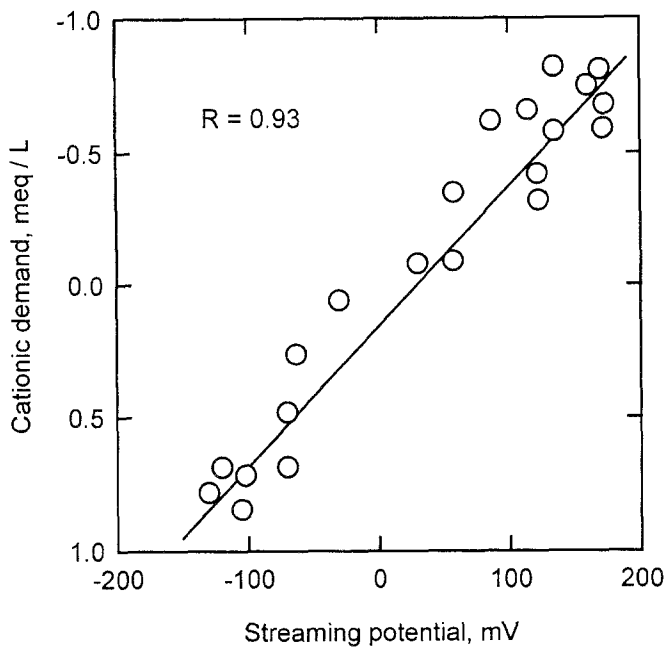
Cationic demand in aqueous media containing discrete fibers, fines, fillers, and other substances directly reflect their surface charge properties; and colloid titration with streaming potential measurement for detecting the accurate



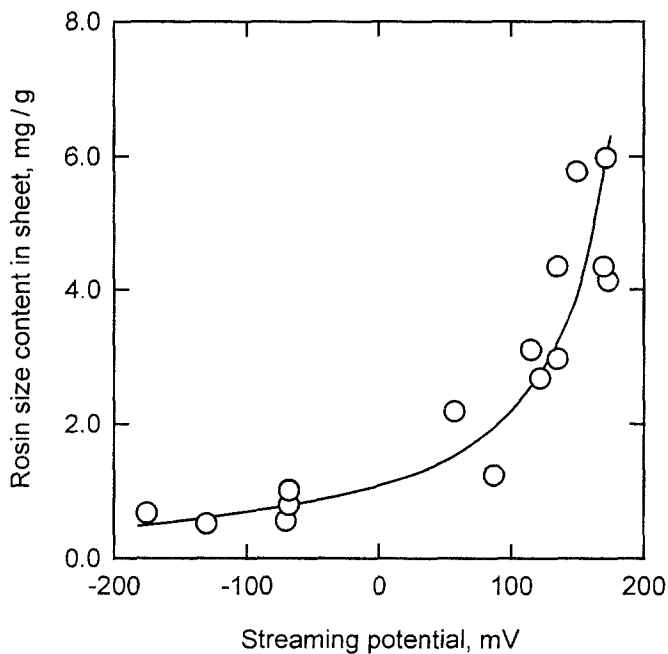
**Fig. 1.** Titrimetric accuracy of the particle charge detector (PCD) system in colloid titration of the mixtures containing  $10^{-4}N$  potassium polyvinyl sulfate (KPVS) and various amounts of aluminum sulfate using  $10^{-4}N$  polydiallyldimethylammonium chloride (PDADMAC)

endpoints has been attempted. It has been reported that the titrimetric accuracy was markedly influenced by the inorganic salts in the systems.<sup>15,21,31</sup>

First,  $10^{-4}N$  KPVS solutions containing various amounts of aluminum sulfate were titrated by  $10^{-4}N$  PDADMAC solutions with the PCD system to evaluate the titrimetric accuracy. Figure 1 shows the titrimetric accuracy calculated from the amounts of KPVS divided by those of PDADMAC required to detect the endpoints with the PCD system. The theoretical isoelectric point must be determined by streaming potential measurements when KPVS reacts with PDADMAC in a 1:1 ratio by ionic association. The titrimetric results corresponded well with the theoretical values until the Al species concentration was lower than 1 mM. Higher concentrations reduced the titrimetric accuracy, and the apparent PDADMAC consumption decreased. This phenomenon has been ascribed to some structural changes in KPVS from linear shapes to random coils by a decrease in the ionic repulsion of each anionic portion whose dissociated acidic groups were interfered with by cations in the systems. The pulp consistency here was 0.15% (w/v), and the addition level of aluminum sulfate was 2% on dry pulp. The concentration of Al ions in pulp suspensions was thus lower than 0.18 mM. The cationic demand determined by colloid titration using KPVS or PDADMAC as a titrant corresponded quite well with the streaming potential values before the polyelectrolyte titration under the various conditions containing both alum and fibers, as shown in Fig. 2. Furthermore, when constant anionic rosin emulsions (1% on dry pulp) were added to the pulp suspensions with various streaming potentials, the

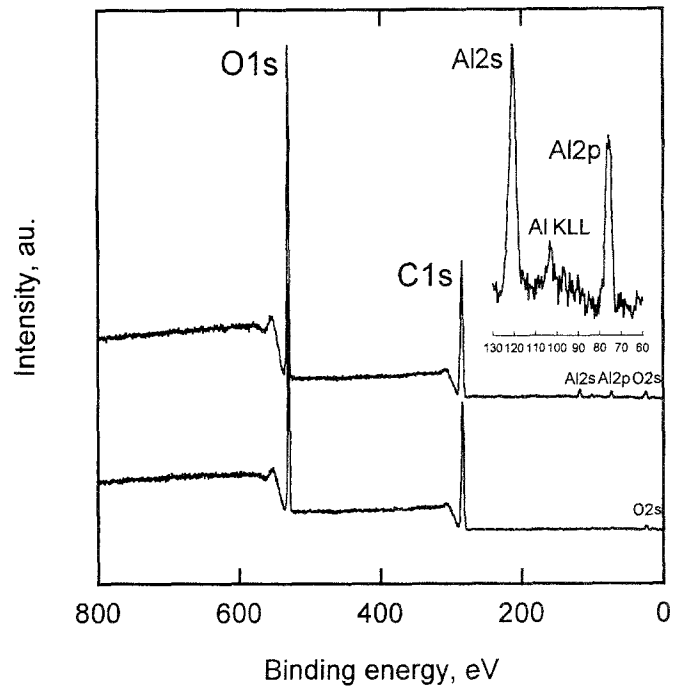


**Fig. 2.** Relation between streaming potentials and cationic demand of pulp suspensions containing aluminum sulfate under various pH conditions



**Fig. 3.** Relation between streaming potentials of aluminum (Al)-containing pulp suspensions just before rosin size addition and rosin size content of pulp sheets prepared after adding emulsion rosin size (1% on dry pulp) to the suspensions

rosin size content of the pulp sheets increased with an increase in the positive streaming potential values of the suspensions, as shown in Fig. 3. These results indicate that the anionic colloidal particles were attracted to the cationized fiber surfaces by ionic interaction in aqueous systems.<sup>23</sup> Thus, it is suggested that the PCD system is a viable option



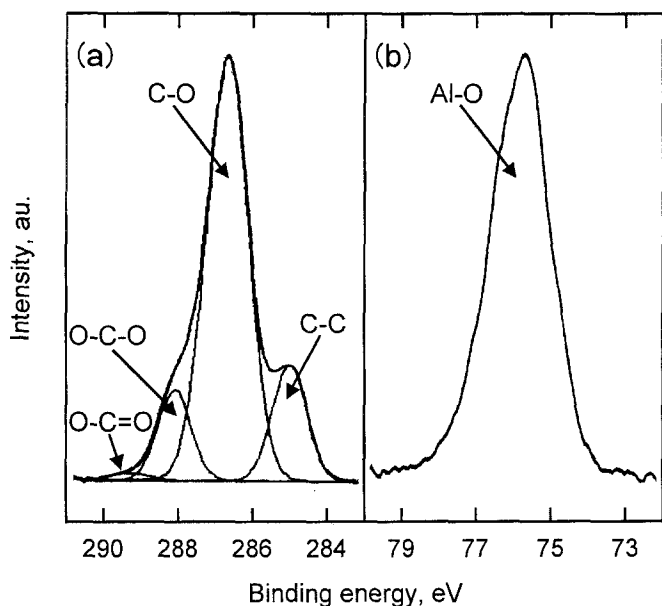
**Fig. 4.** X-ray photoelectron spectroscopy (XPS) survey spectra of pulp sheets prepared with (top spectrum) or without (bottom spectrum) aluminum sulfate. **Inset** Magnified Al signals of the pulp sheet containing aluminum components

for determining the streaming potentials closely related to the surface charges of the pulp fibers in colloidal systems containing a small concentration of Al ions.

#### XPS analysis of pulp sheets containing Al components

Analysis by XPS offers particular information for atomic compositions of the thin-layer regions on solid surfaces. Figure 4 shows the typical XPS spectra of pulp sheets prepared from pulp mixtures with or without aluminum sulfate. Clear signals due to the XPS C1s and O1s of cellulosic pulp fibers were detected at survey spectra of the pulp sheets. Al2s and Al2p signals were measured on the survey spectrum of an alum-added pulp sheet. The Al KLL weak band between Al2s and Al2p signals was an Auger peak of the Al element. Various atomic peaks were then determined independently, and the intensity at each BE corresponds to concentrations of the elements present in the detectable regions. Hence XPS is convenient for qualitative and quantitative elemental analyses of the solid surfaces and does not require troublesome pretreatment of the samples.

Figure 5 shows narrow scanned spectra at focused BE ranges of each element and deconvolution patterns of the C1s signal of the alum-added pulp sheet as shown in Fig. 4 by a high-resolution analytical mode. A curve-fitting program with Gaussian and Lorentz's functions was supplied with the XPS system. The carbon atoms in the pulp sheet were divided into four groups based on their chemical shifts,<sup>24-26</sup> as shown in Fig. 5a. Peaks due to the C—O band at 286.7 eV of BE and the O—C—O band at 288.1 eV of BE

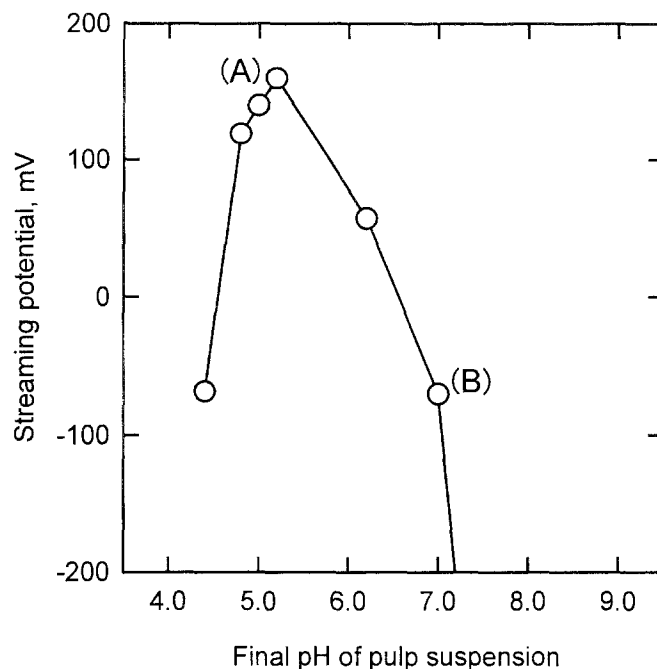


**Fig. 5.** XPS narrow scanned spectra of pulp sheets prepared with aluminum sulfate (2% on dry pulp). **a** C1s signal. **b** Al2p signal

originate from the cellulosic fibers; and a slight band of O—C=O derived from a carboxyl group at 289.3 eV of BE was detected. A peak of the C—C band at 285.0 eV of BE was absent in cellulose and hemicellulose, although even in bleached kraft pulps its presence was reported as residual lignin components or wood extracts.<sup>24,26,32</sup> The Al components on pulp fiber surfaces were analyzed with regard to the peak of the Al2p signal, as shown in Fig. 5b, because the peak of the Al2p signal was sharper and more sensitive than that of Al2s on each baseline. Most Al2p bands measured in this study were determined at about 75.5–76.0 eV of BE under the various conditions for preparing the pulp sheets. Chemical shifts of unoxidized Al are about 73–74 eV of BE. Thus, the chemical structures of Al components on the surfaces of the pulp sheets must mainly be Al—O forms, at least during the XPS measurement. Detailed chemical structures of the Al components on the fiber surfaces have not been clarified. The existence of Al components on the fiber surfaces, called “surface aluminum content” herein, was estimated by the atomic ratios calculated from the relative peak areas of Al2p against C1s for both quantitative and mapping analyses for reducing the inevitable influence of the surface roughness of the sheet samples.

#### Relations between fiber charge characteristics and Al content of pulp sheets

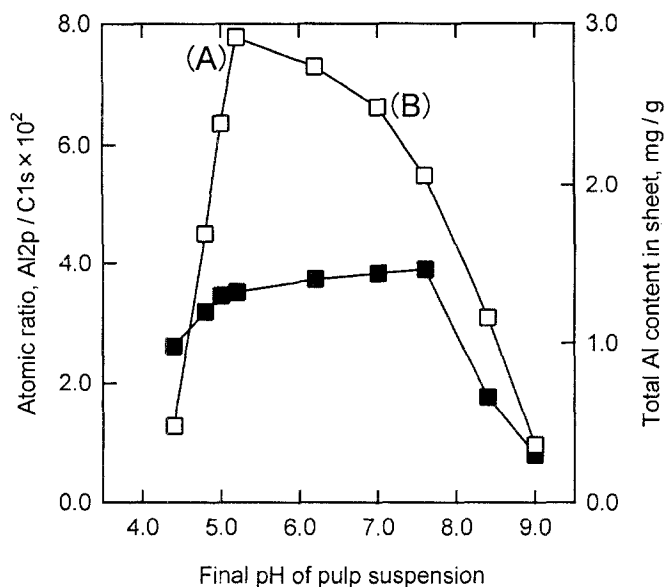
Aluminum ions originating from aluminum sulfate added to the paper stock have some impact on drainage and retention behavior because they presumably affect the charge properties of the fiber surfaces at the wet end. Figure 6 shows the streaming potentials of pulp suspensions at various pH levels controlled by adding a dilute NaOH solution after 3 min of aluminum sulfate addition (2% on dry pulp). This pulp was washed beforehand with a dilute HCl solu-



**Fig. 6.** Streaming potentials of pulp suspensions containing aluminum sulfate (2% on dry pulp) at various pH levels. Symbols (A) and (B) in this figure are marked for Fig. 10

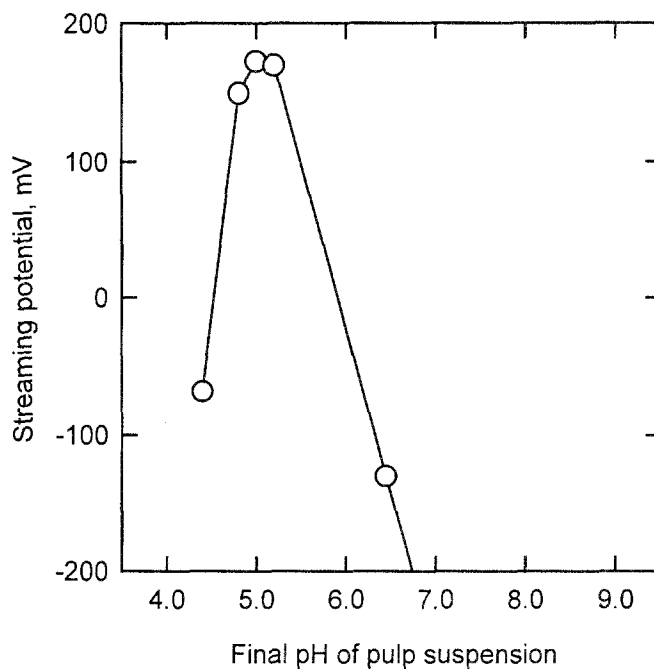
tion to remove dissolved anionic trash, and all carboxyl groups of the pulp fibers were converted to a free-acid type (pulp—COOH).<sup>29</sup> The initial streaming potential of the pulp suspension prepared using deionized water was negative: about  $-900$  mV at pH 5 (data not shown); aluminum sulfate addition drastically increased it to about  $-70$  mV at pH 4.4. The positive regions appeared on the pulp fibers owing to adsorption of Al cations on the fiber surfaces. By adding a dilute NaOH solution in limited amounts to the suspension, the fiber surface charge converts from slightly negative to positive in the narrow range of around pH 5. Moreover, rapid decreases of streaming potentials by further alkali additions were observed at  $> \text{pH } 5.2$ . In general, positive charges due to the cations present on the solid surfaces were immediately consumed according to the adsorption of  $\text{OH}^-$  ions via simple ionic interaction. The Al species in the pulp suspensions, however, distinctly contribute to cationization of the fiber surfaces by adding  $\text{OH}^-$  ions. Maximum streaming potential value (ca.  $+160$  mV) was detected when the  $\text{OH}^-/\text{Al}$  molar ratio in the system was about 2.5. The optimum neutralization of Al solutions obtained by titratable charge measurements under equilibrium conditions was reported to be about 1.5–2.0.<sup>2,5,12</sup> These differences probably arise from the carboxyl groups of the pulp fibers, which act as an acid in the neutralization procedures.

Next, the Al components adsorbed on the pulp fibers were determined quantitatively by XPS and XFA. Figure 7 shows the atomic ratios of the Al2p peak area divided by that of C1s on the fiber surfaces and total amounts of Al retained in the pulp sheets prepared with aluminum sulfate under various pH conditions. During the XPS analysis the atomic concentration is sensitively quantified by detecting



**Fig. 7.** Surface (open squares) and total (filled squares) Al contents of pulp sheets prepared from the pulp suspensions as shown in Fig. 6. Symbols (A) and (B) in this figure are marked for Fig. 10

the photoelectrons that escape only from the surfaces of the samples because the photoelectrons generated in the core of the sample are inevitably disturbed by the other portions of the samples. Therefore, detectable regions of the specimen are limited to the surfaces (< ca. 10 nm). On the other hand, during the XFA measurement the elemental content of the samples is determined quantitatively by the intensity of the characteristic X-rays radiated from the samples; and most of the composition of both surfaces and the insides of the samples are detectable because of the high permeability of X-rays. In the pulp mixtures at around pH 4.4, the total Al retention was about 31% of the amount of aluminum sulfate added, and the surface aluminum content was determined to be low. It was reported that these aluminum species were adsorbed on the fibers at the carboxyl groups of the pulp,<sup>10,23</sup> and an ion-exchange reaction occurred stoichiometrically under equilibrium conditions.<sup>35</sup> The carboxyl content of the pulp fibers is 0.06 mmol/g of the dry pulp; and about 60% of their carboxyl groups were plausibly converted to pulp-COOAl structures by reaction with the Al species. The pH increase of the pulp suspension by the alkali additions brought about a marked increase in the surface Al content of the pulp sheets compared with the total Al content of the sheets. These results strongly suggest that the residual Al species in the suspensions were preferentially adsorbed on the fiber surfaces, and additionally adsorbed Al components improved the fiber charge characteristics, as shown in Fig. 6. Evidently, streaming potentials of the suspension reached a maximum value at the highest surface Al content of the sheets, as shown in Figs. 6 and 7. Further supplements of OH<sup>-</sup> ions caused a gradual increase in the total Al content of the pulp sheets, whereas the surface Al content and the fiber charges clearly decreased in the pH range 5.2–7.6. In this pH range it was assumed that OH<sup>-</sup> ions added to the suspensions consumed the positive



**Fig. 8.** Streaming potentials of pulp suspensions containing aluminum sulfate (2% on dry pulp) at various pH levels. Aluminum sulfate was added after the prescribed alkali treatments of the pulp suspensions

charges of the fiber surfaces and promoted the formation of aluminum flocs > 10 nm in diameter; the surface Al content then seemed to decrease apparently because XPS determined the Al elements only on the surfaces of the Al flocs. Excess additions of alkali cleaved certain interactions between the Al flocs and the pulp fibers.<sup>9,10,23</sup> Accordingly, the Al components on the fiber surfaces and the total Al content of the sheets decreased quickly at > pH 7.6. Figures 8 and 9 show that the streaming potentials of the pulp suspensions and the Al content of the sheets are similar to those shown in Figs. 6 and 7, respectively, when aluminum sulfate was added to the suspension after adding the prescribed amounts of alkali solution. The degree of neutralization when the streaming potential reached a maximum was shifted from 2.5 to 2.0, but the fundamental relation between the streaming potentials and the surface Al content was maintained. In brief, the charge properties of the pulp fibers at the wet end correlated roughly with the surface Al content of the pulp sheets under the acidic conditions of the pulp suspensions containing aluminum sulfate.

#### Distribution characteristics of Al components on fiber surfaces

Fiber charge characteristics of the pulp suspensions corresponded closely with the surface Al content of the sheets < pH 5.2. Further alkali addition rapidly reduced the streaming potential values of the systems compared with a gradual decrease of the surface Al content, as shown in Figs. 6 and 7 and Figs. 8 and 9. Two conspicuous sheets (A and B in Figs. 6 and 7) were determined by two-dimensional XPS

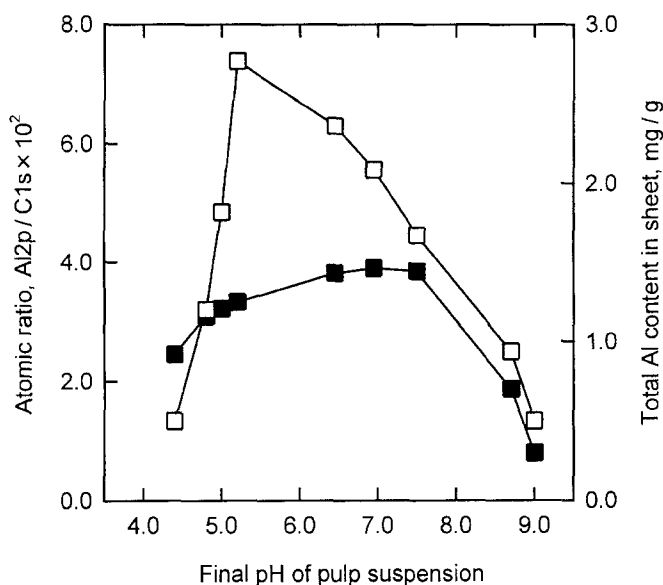
analysis for more detailed characterization of the fiber surfaces. Figure 10 shows the mapping images of the Al<sub>2p</sub> signal against that of C1s after each baseline correction. Sheet A has the most positive streaming potential value and the highest surface Al content. Sheet B shows a negative charge for the fibers despite a slight decrease in the surface Al content, in contrast to that of sheet A. As a result, the Al components on these sheets were roughly uniform in sheet A, in contrast to the obvious heterogeneous distribution in sheet B. Such localization of the Al components on the fiber surfaces suggests rapid coagulation of the aluminum hydroxide formed by the reaction with

OH<sup>-</sup> ions added to the suspension above the optimum. Therefore, it is indicated that the adsorption behavior of Al species at around pH 5 implies the formation of noncoagulable and cationic Al complexes on the fiber surfaces. Excessive alkali supplements, however, spoil the effective Al cations and produce only the useless nonionic Al flocs.

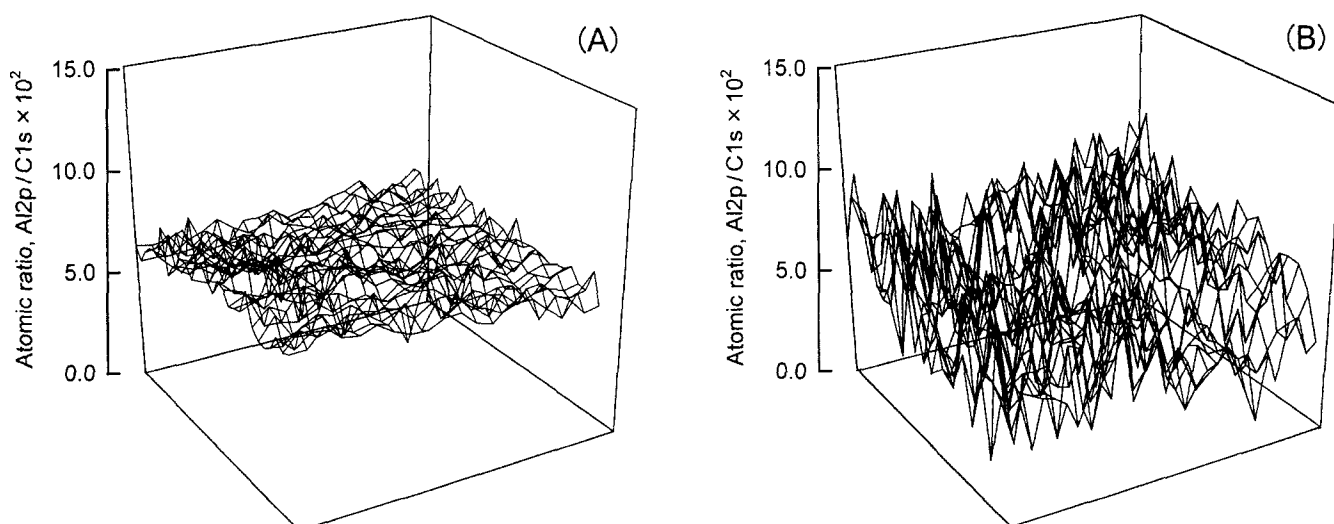
During practical papermaking processes, many ionic interfering substances are present in the paper furnish. These complicated situations must be investigated to elucidate the factors affecting the final performance of Al compounds at the wet end.

## Conclusions

Fiber charge characteristics of pulp suspensions containing aluminum sulfate were determined by the PCD system; and the adsorption behavior of Al components of the pulp fibers were investigated quantitatively by two X-ray elementary analyses – XPS and XFA – to elucidate why the moderate alkali supplements that generally consume the positive charges improve the performance of Al compounds as papermaking additives. The Al species added to the pulp suspensions were adsorbed indistinguishably onto and into the pulp fibers under acidic conditions. The optimum alkali additions stimulated the preferential and uniform adsorption of Al cations on the fiber surfaces at around pH 5. Excess neutralization procedures produced nonionic aluminum flocs on the fibers and drastically reduced the streaming potentials of the suspensions, although the total Al retention was constant. It was revealed that the hydroxyl ions in Al-containing pulp suspensions probably play an important role in promoting adsorption of effectual Al cations onto the fiber surfaces accessible for the wet-end additives and other substances, rather than the structural



**Fig. 9.** Surface (*open squares*) and total (*filled squares*) Al contents of pulp sheets prepared from the pulp suspensions as shown in Fig. 8



**Fig. 10.** Two-dimensional XPS images of the surfaces of pulp sheets prepared with aluminum sulfate (2% on dry pulp) at pH 5.2 (**A**) and pH 7.0 (**B**) adjusted by adding a dilute alkali solution, as shown

in Figs. 6 and 7. These images are partially illustrated from the whole XPS mapping areas, and each graphic area corresponds to  $0.15 \times 0.15$  mm

changes and charge variation of the Al complexes themselves.

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